Soft x-ray spectroscopy of Dy, Er and Tm ions excited in laser-produced plasmas

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2017 Source Workshop, 6th – 8th November 2017, Dublin, Ireland

Outline

- Introduction
  - Motivation.
- Experiment
  - Setup & power density dependence of spectra.
- Calculations and identifications
  - Ag -, Pd - and Rh - like features in $\Delta n = 0$ features.
  - Sr - like isoelectronic study.
  - $\Delta n = 1$ transitions: importance of satellite transitions.
- Summary

Lanthanide ions: Short λ sources

BEUV Lithography

- Emission @ 6.X nm to match reflectivity of La/B_4C MLMs?
- Moving to higher Z, open 4d subshell Sn UTA @ 13.5 nm moves to shorter λ:

$$\lambda = a(Z - s)^{-b}/R_\infty$$

(H. Ohashi *et al.* , Appl. Phys. Lett. **104**, 234107 (2014))

- **Gd (Z = 64)** and **Tb (Z = 65)** ions: Intense UTAs emitted around 6.5/6.7 nm (Δλ ~ 0.1 nm).

(Churilov *et al.* Phys. Scr. **80**, 045303 (2009))

Problem: Maximum CE (published) = **0.8 %**
(K. Yoshida *et al*. Appl. Phys. Express. **7**, 086202 (2014))

Application: Sources for metrology?

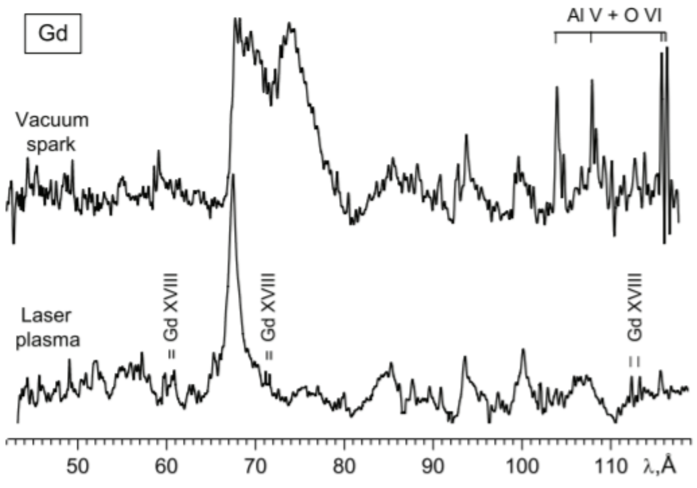


Figure 1. Spectra of gadolinium ions excited in the vacuum spark (upper trace) and in the laser-produced plasma (bottom trace).

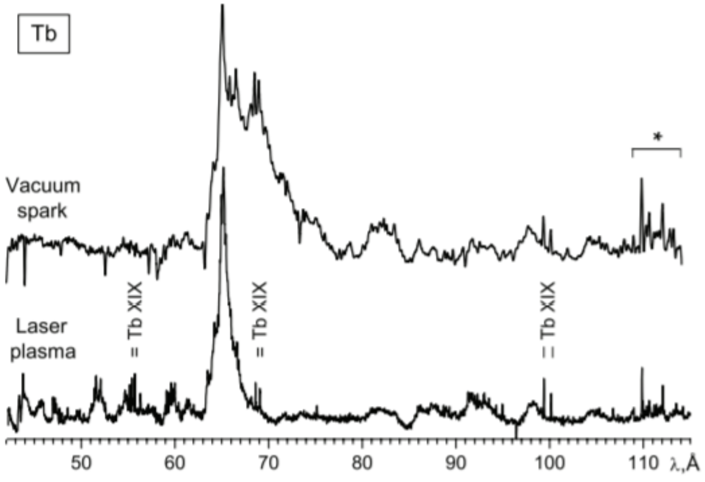
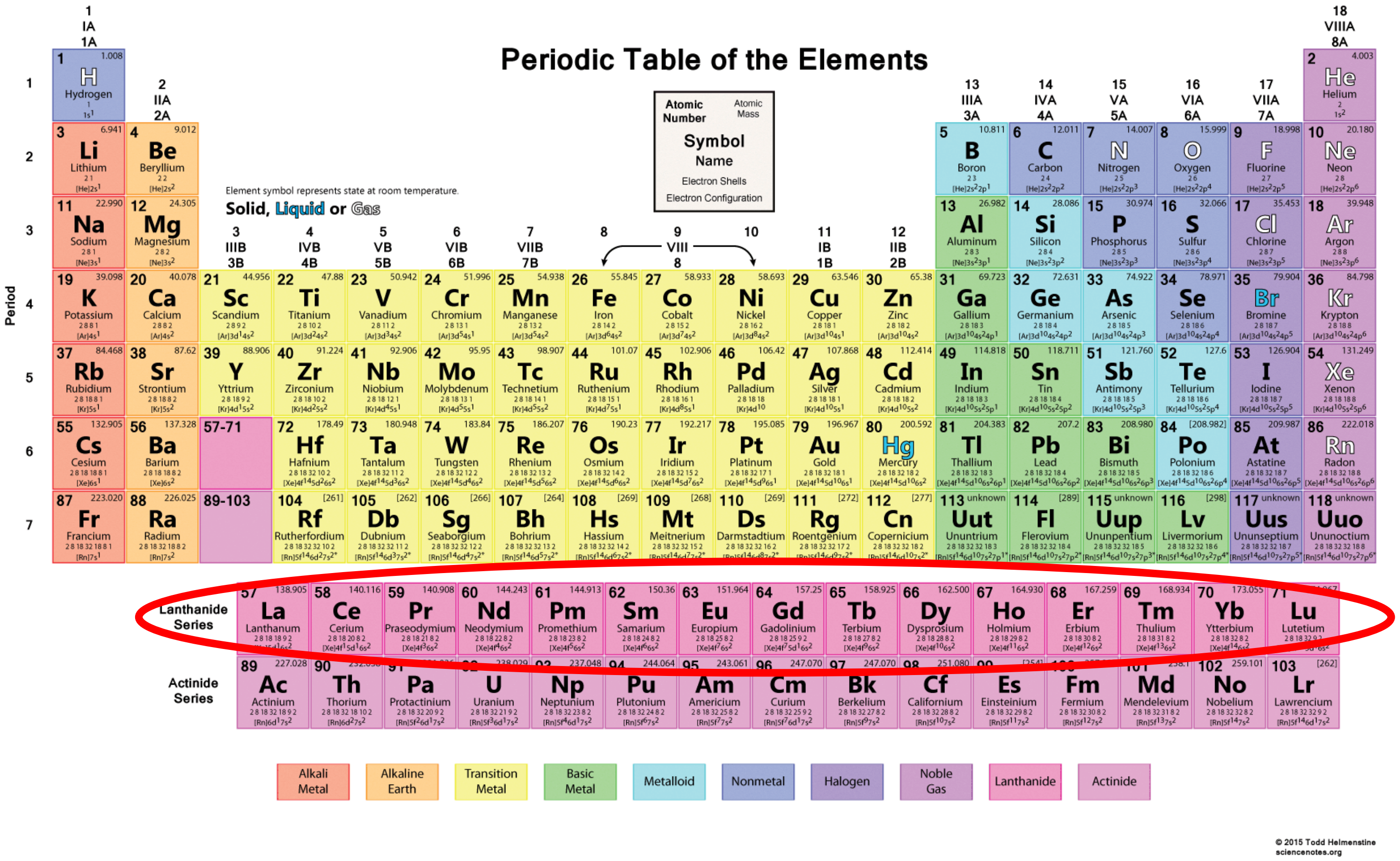


Figure 2. Spectra of terbium ions excited in the vacuum spark (upper trace) and in the laser-produced plasma (bottom trace). *, $4f^2$–4f5d transition array in Tb XVIII classified in the present work.

Where are we?



Periodic Table of the Elements
Atomic Number
Atomic Mass
Symbol
Name
Electron Shells
Electron Configuration
Element symbol represents state at room temperature.
Solid, Liquid or Gas
Lanthanide Series
Actinide Series
Alkali Metal
Alkaline Earth
Transition Metal
Basic Metal
Metalloid
Nonmetal
Halogen
Noble Gas
Lanthanide
Actinide
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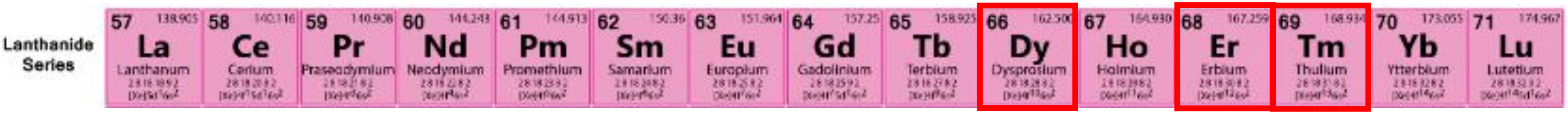
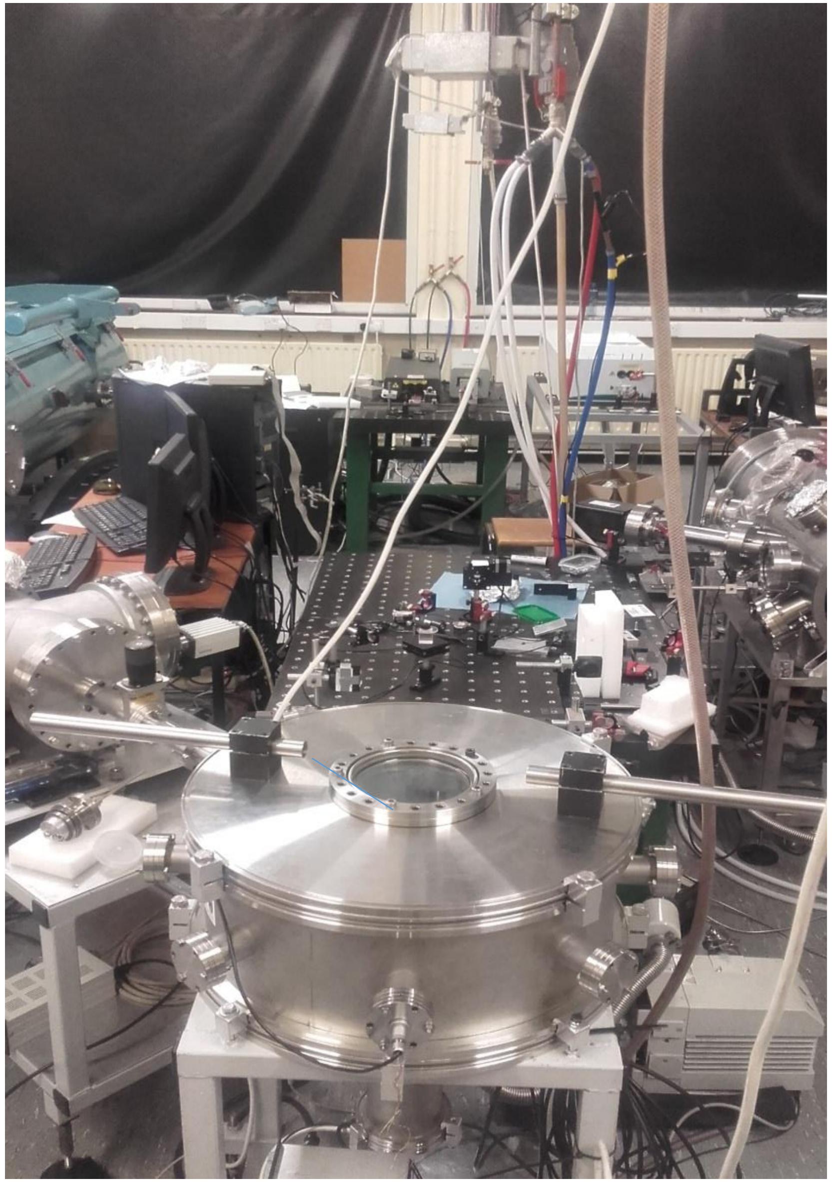


Lanthanide Series
57 La Lanthanum
58 Ce Cerium
59 Pr Praseodymium
60 Nd Neodymium
61 Pm Promethium
62 Sm Samarium
63 Eu Europium
64 Gd Gadolinium
65 Tb Terbium
66 Dy Dysprosium
67 Ho Holmium
68 Er Erbium
69 Tm Thulium
70 Yb Ytterbium
71 Lu Lutetium

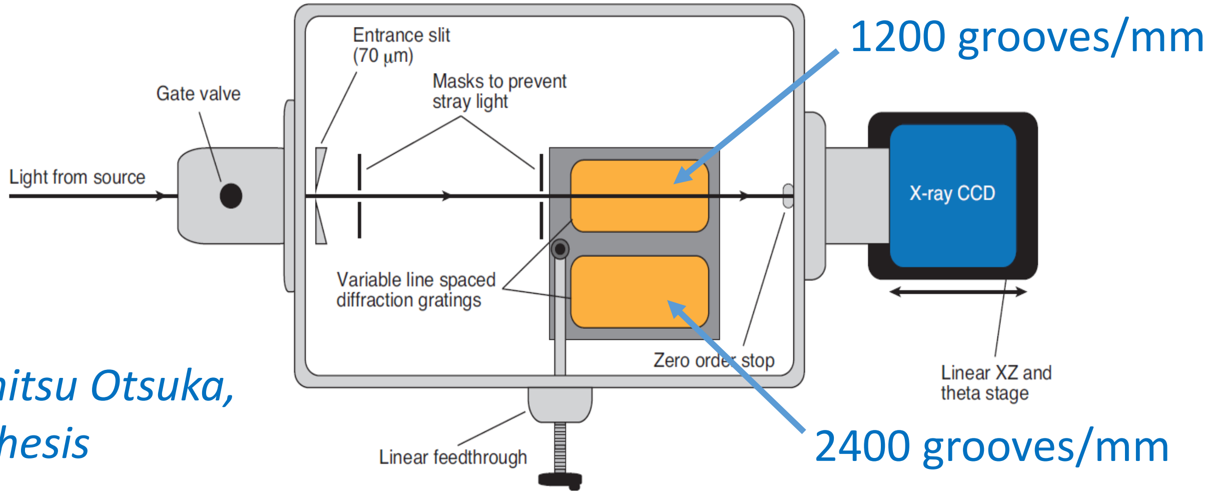
LPP experiments



Experimental parameters

- 7 ns FWHM Nd:YAG laser
- Max energy ≈ 750 mJ
- Spot size ≈ 45 µm
- Power densities: $(1.3 \; - 6.6) \text{ x } 10^{12} \text{ W/cm}^2$

0.25 m flat-field grazing incidence spectrograph

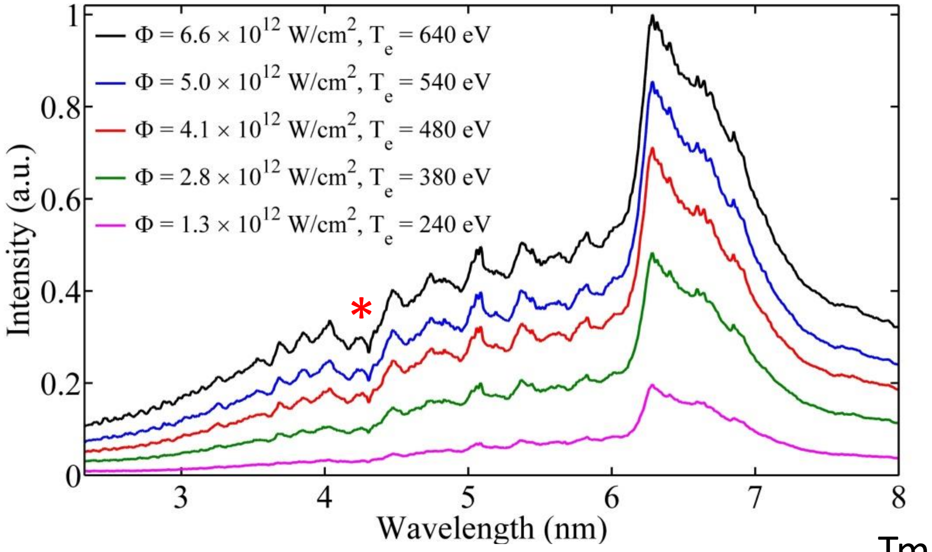


Takamitsu Otsuka,
PhD Thesis

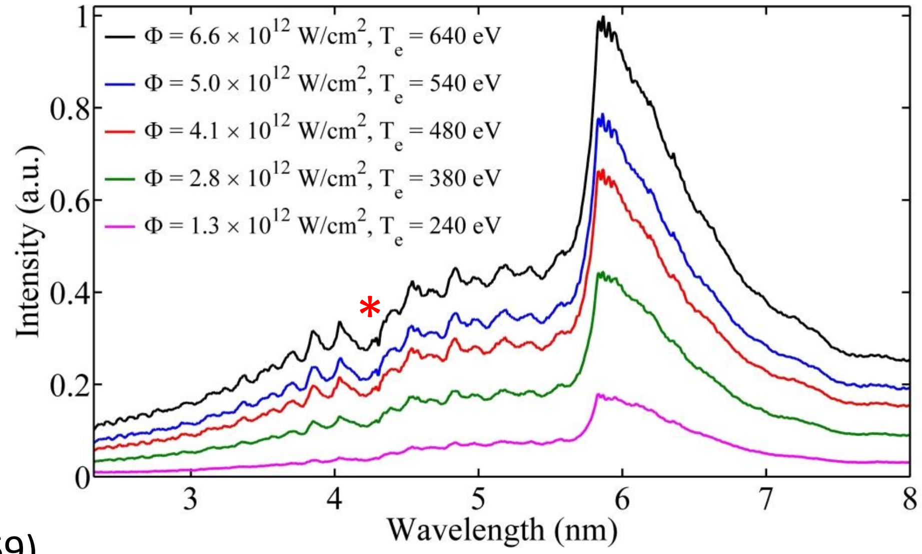
Spectral resolution: 0.03 nm @ 6 nm
Wavelength uncertainty: 0.003 nm

Power density dependence

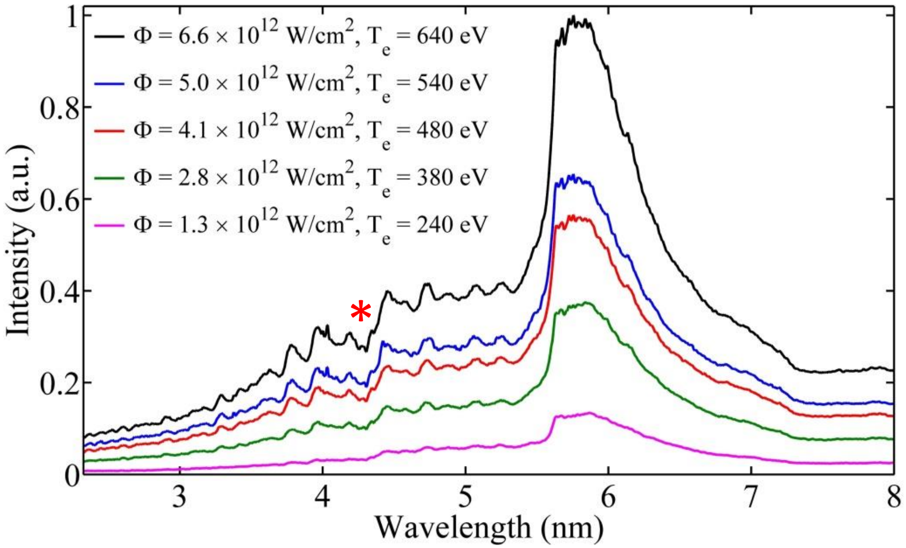
Dy (Z = 66)



Er (Z = 68)



Tm (Z = 69)



*<u>**Carbon K-edge absorption**</u>

Theoretical Calculations

Cowan code

- Hartree-Fock with configuration interaction (HFCI).
- Relativistic and correlation correction modes used.
- Ability to scale Slater integrals to account for CI from missing configurations.

Configurations

4d subshell

Lower: $4d^k$ ($+ 4d^{k-1}5s$)

Upper: $4d^{k-1}np$ (n = 5 - 7), $4d^{k-1}nf$ (n = 4 - 7) and $4p^5 4d^{k+1}$ ($+ 4d^{k-2}5s5p$)

4f subshell (resonance)

Lower: $4d^{10}4f^m$

Upper: $4d^{10}4f^{m-1}5d$, 5g, 6d and 6g

4f subshell (satellite)

Lower: $4d^{10}4f^{m-1}5s$

Upper: $4d^{10}4f^{m-2}5s5d$, 5g, 6d and 6g

Scaling factors

0.85(F^k, G^k, R^k) integrals and *ab initio* ζ integrals.

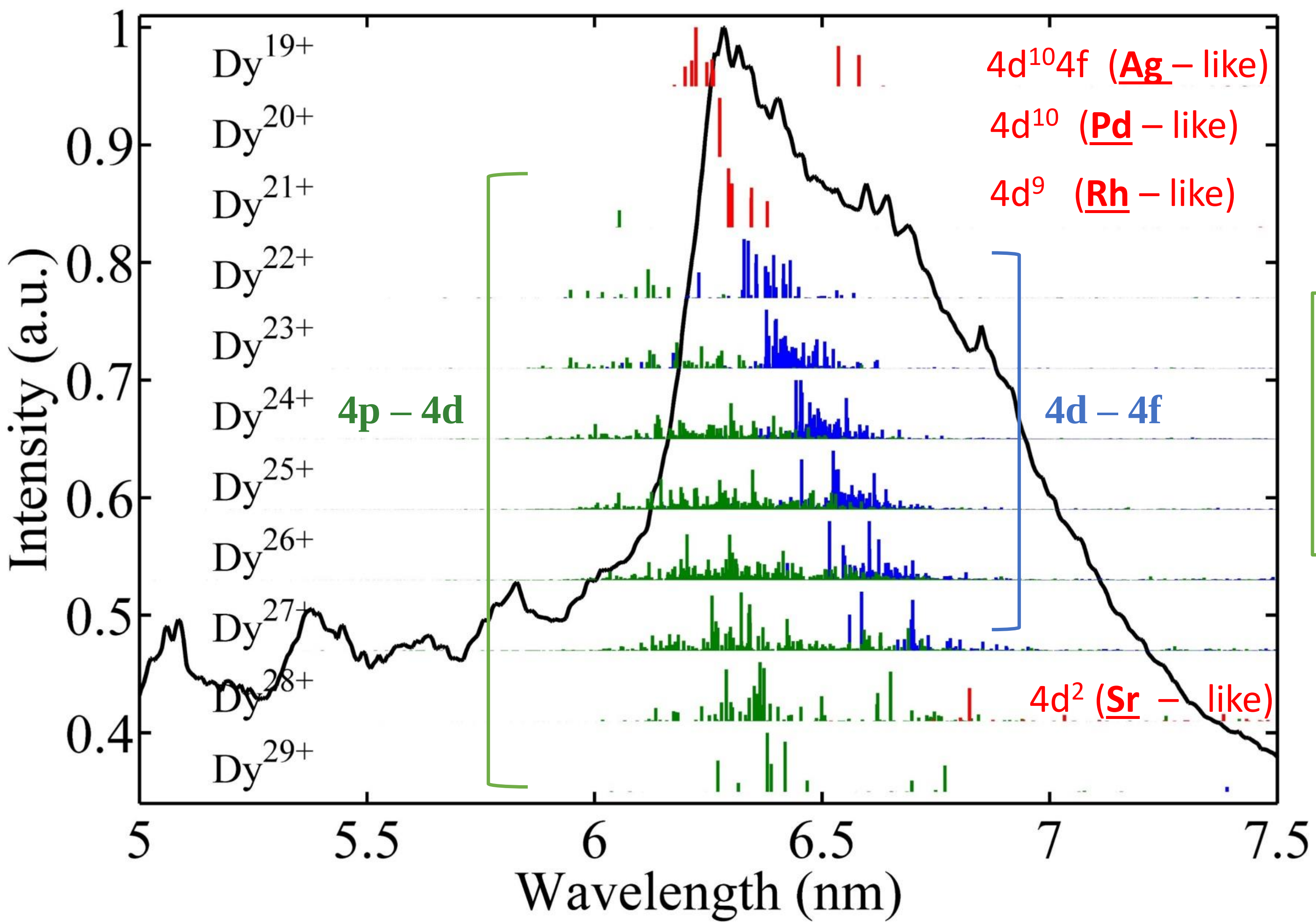
Flexible Atomic Code

- Fully relativistic, *ab initio* approach.
- Solves the Dirac equation in a model potential.
- Suitable for high-Z, highly-charged ions.
- Calculated Ag -, Pd -, Rh - and Sr - like spectra.

Same configurations used as Cowan code

Comparison is very useful when identifying features!!

Δn = 0, n = 4 – n = 4 UTA: Dy



Subvalence 4p – 4d excitation weak - include to obtain accurate wavefunctions (configuration interaction)

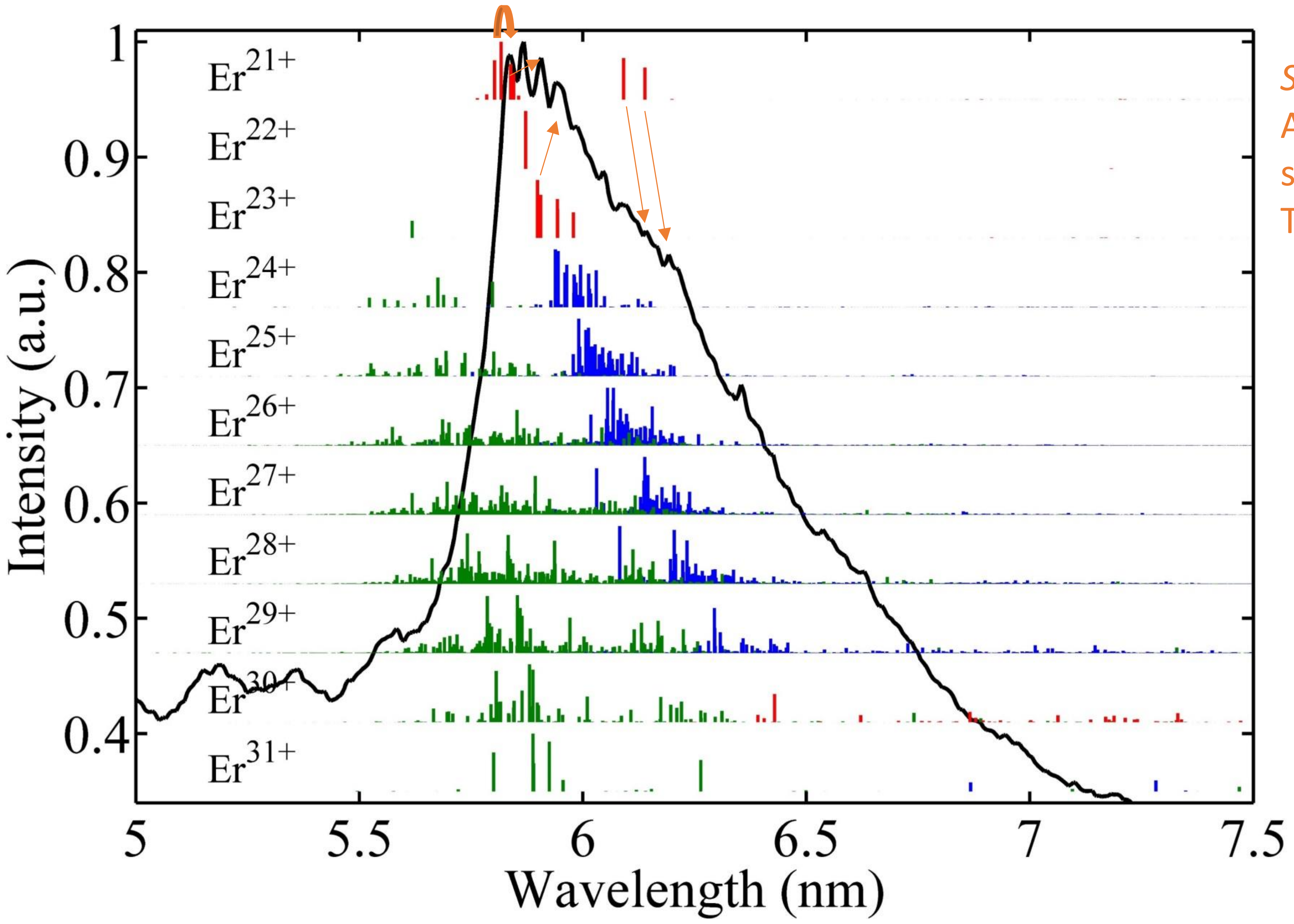
Case study: Ag – like Dy

Transition	Previous (nm) λ_{Exp}	Previous (nm) λ_{Theor}	Current (nm) λ_{Exp}	Current (nm) λ_{Cowan}	Current (nm) λ_{FAC}	$\Delta\lambda$ (nm) |Exp|	$\Delta\lambda$ (nm) Exp_{prev} – FAC
$4d^{10}4f\ ^2F_{5/2} - 4d^94f(^3H)4f\ ^2G_{7/2}$	6.6437[a],6.641[b]	6.5869[c],6.398[b]	6.642	6.5807	6.4847	0.0017	0.1590
$4d^{10}4f\ ^2F_{7/2} - 4d^94f(^3H)4f\ ^2G_{9/2}$	6.5967[a],6.595[b]	6.5420[c],6.349[b]	6.597	6.5360	6.4348	0.0003	0.1619
$4d^{10}4f\ ^2F_{5/2} - 4d^94f(^3H)4f\ ^2F_{5/2}$	6.2535[c*]	6.2052[c]		6.1992	6.0589		0.1946
$4d^{10}4f\ ^2F_{7/2} - 4d^94f(^3H)4f\ ^2F_{7/2}$	6.2587[c]	6.2284[c]		6.2227	6.0814		0.1773
$4d^{10}4f\ ^2F_{5/2} - 4d^94f(^3F)4f\ ^2D_{3/2}$	6.3125[a]	6.2652[c]	6.315[bl]	6.2583	6.1199	0.0025	0.1926
$4d^{10}4f\ ^2F_{7/2} - 4d^94f(^3F)4f\ ^2D_{5/2}$	6.3163[c*]	6.2679[c]	6.315[bl]	6.2614	6.1198	0.0013	0.1965
$4d^{10}4f\ ^2F_{5/2} - 4d^{10}5g\ ^2G_{7/2}$	5.0586[a]		5.061	5.0577	5.0779	0.0024	- 0.0193
$4d^{10}4f\ ^2F_{7/2} - 4d^{10}5g\ ^2G_{9/2}$	5.0887[a]		5.085	5.0869	5.1072	0.0037	- 0.0185

± 0.0035
(Combined uncertainty)

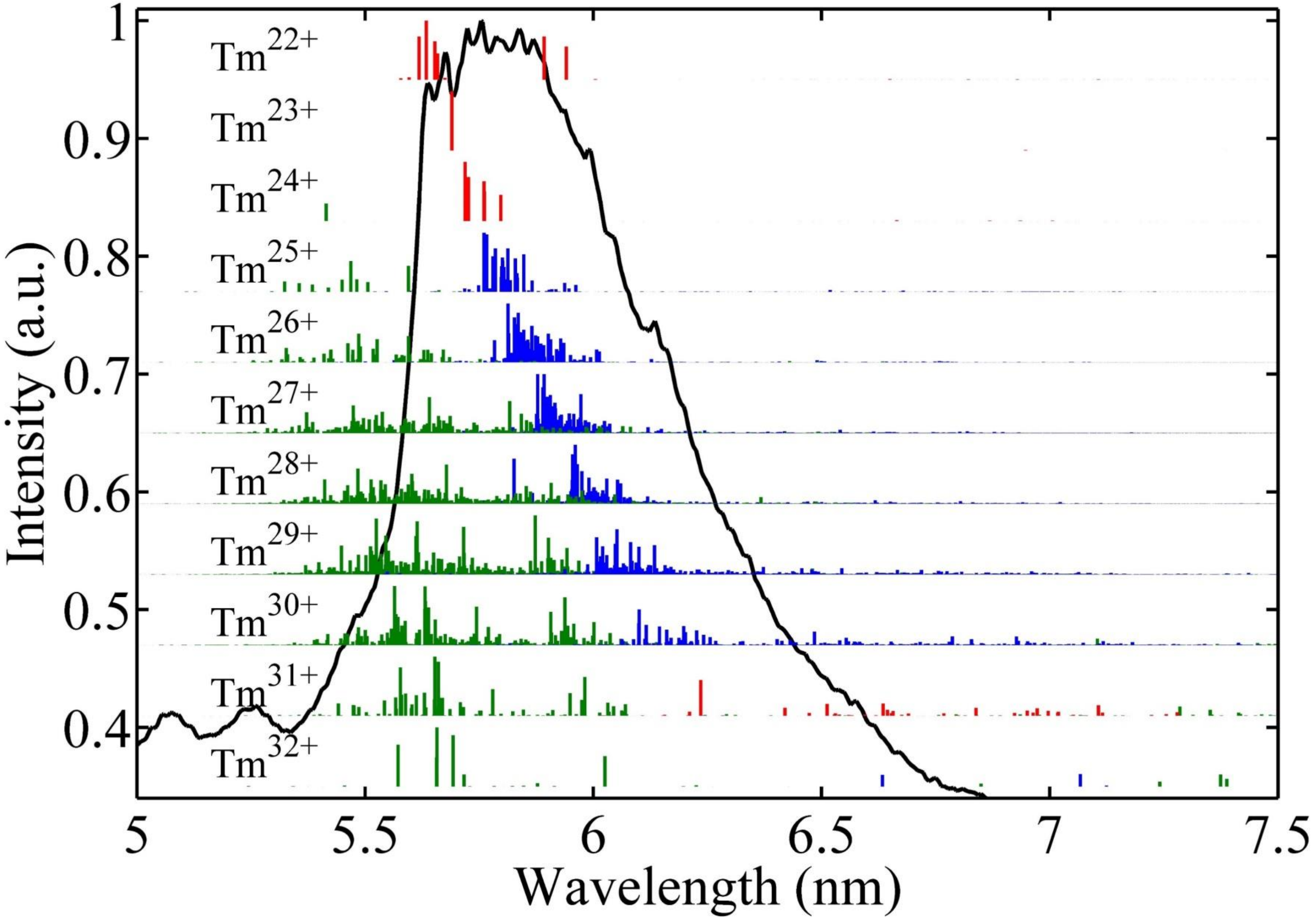
[a]Sugar J and Kaufman V 1981 *Phys. Scr.* **24** 742
[b]Suzuki C, *et al* 2015 *J. Phys. B. At. Mol. Opt. Phys.* **48** 144012
[c]Sugar J, Rowan W L and Kaufman V 1993 *J. Opt. Soc. Am B* **10** 1321
*Semi-empirical values predicted using a fitted curve to the differences between the observed and calculated transition energies.
[bl]Blended with other emission lines: peak wavelength of feature is reported.

Δn = 0, n = 4 – n = 4 UTA: Er

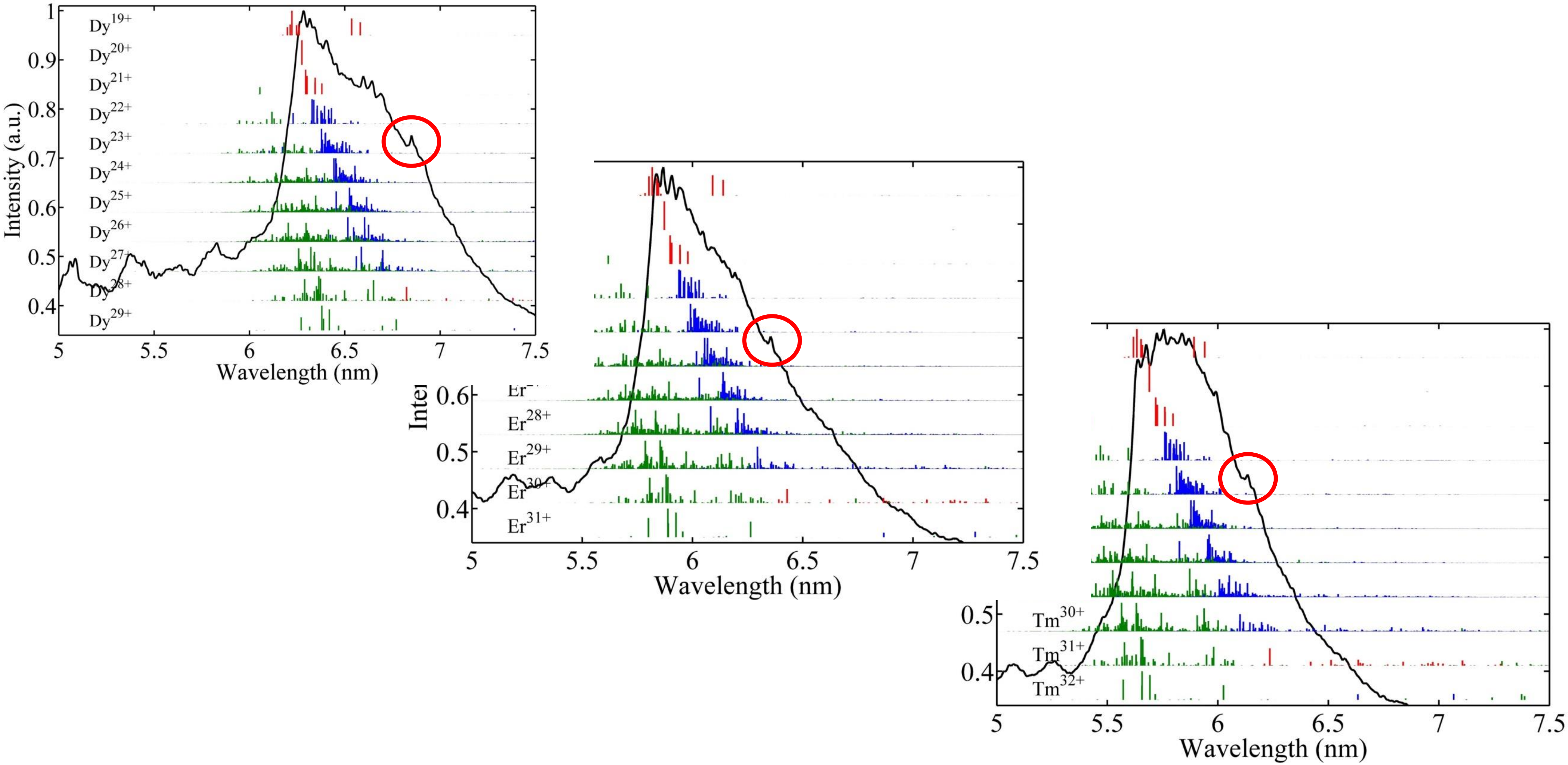


Sugar, Kaufman and Rowan:
Ag -, Pd - and Rh - like high - Z spectra studied in the TEXT Tokamak.

$\Delta n = 0$, n = 4 – n = 4 UTA: Tm

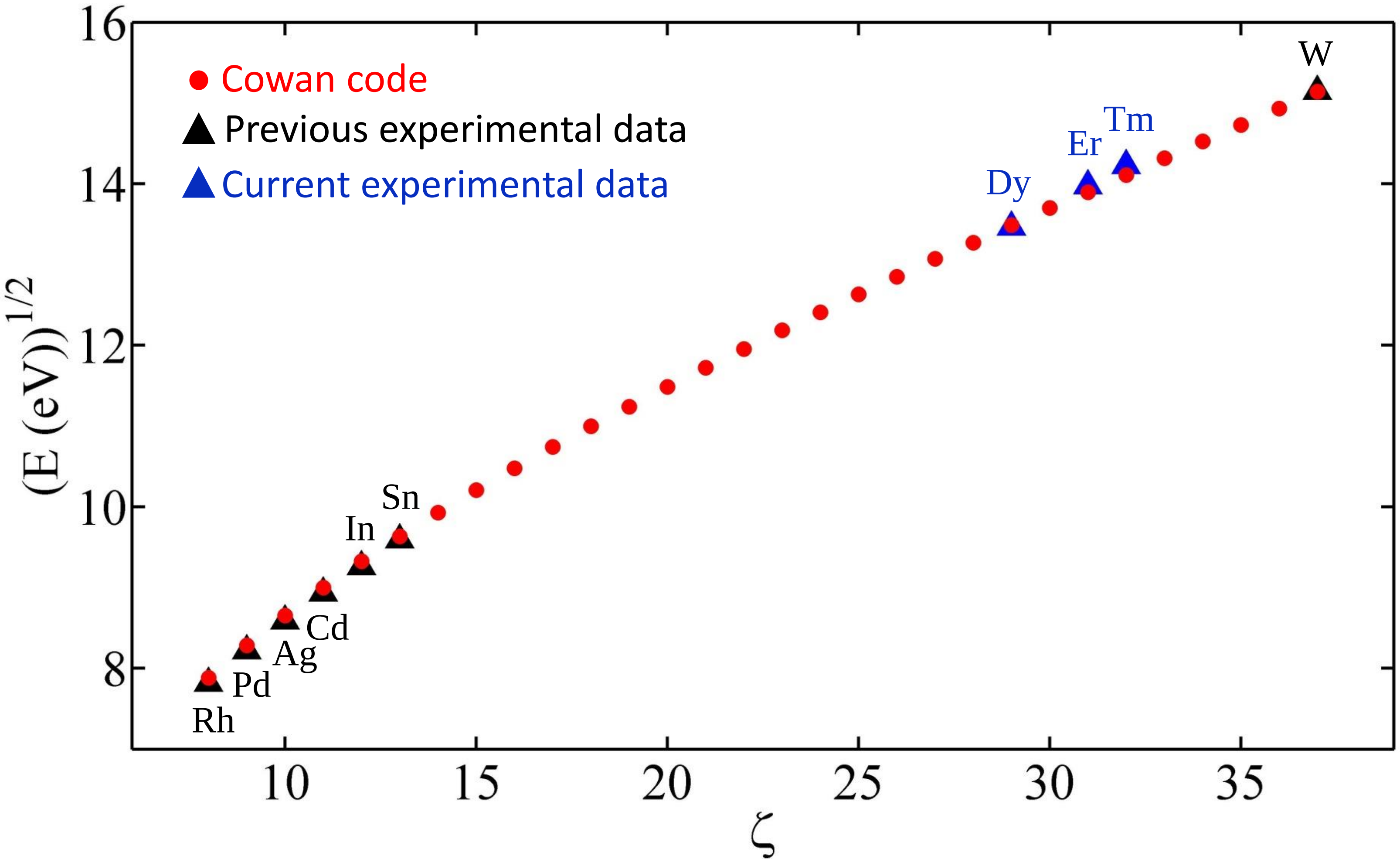


Did you notice…….?

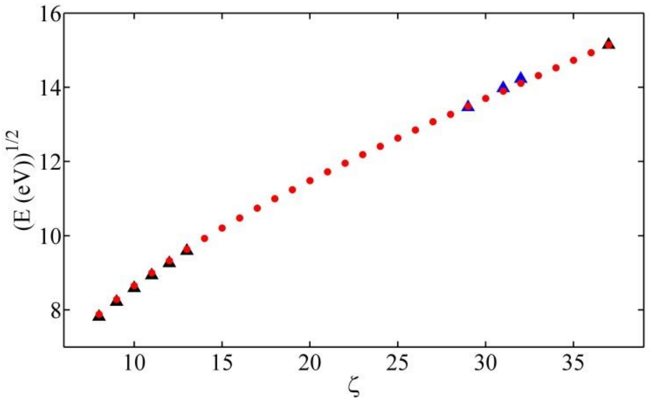


Isoelectronic extrapolation: $4d^2\ {}^3F_2$- $4d4f\ {}^3G_3$

Edlén plot: $\mathrm{E(eV)}^{1/2}$ versus $\zeta = Z - (N-1)$ → Accurately interpolate/extrapolate wavenumbers.



Isoelectronic extrapolation: $4d^2\ {}^3F_2$- $4d4f\ {}^3G_3$



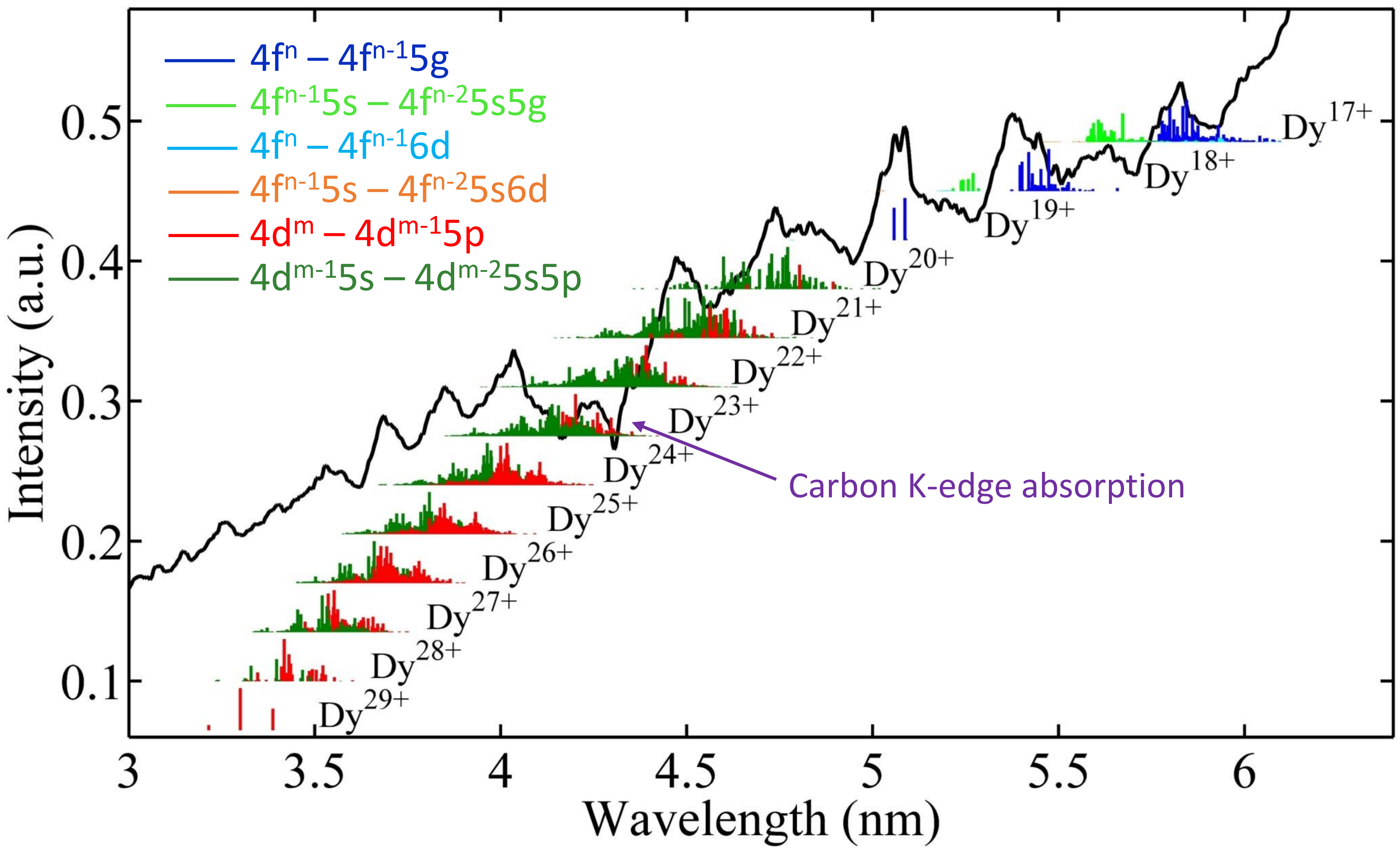
Ion	λ_{Exp} (nm)	Leading eigenvector component LS	jj	λ_{Cowan} (nm)	$gA/\Sigma gA$	$\Delta\lambda$ (Exp – Cowan)	λ_{FAC} (nm)	$\Delta\lambda$ (Exp – FAC)
Dy^{28+}	6.849	44% $4d(^2D)4f\ ^3G_3$	67% $4d4f(3/2,5/2)_3$	6.8242	0.30	**0.025**	6.7041	0.145
Er^{30+}	6.354	64% $4d(^2D)4f\ ^3G_3$	57% $4d4f(3/2,5/2)_3$	6.4289	0.21	- 0.075	6.3142	0.040
Tm^{31+}	6.135	59% $4d(^2D)4f\ ^3G_3$	72% $4d4f(3/2,5/2)_3$	6.2353	0.21	- 0.100	6.1255	**0.009**

Reasons for uncertainty:

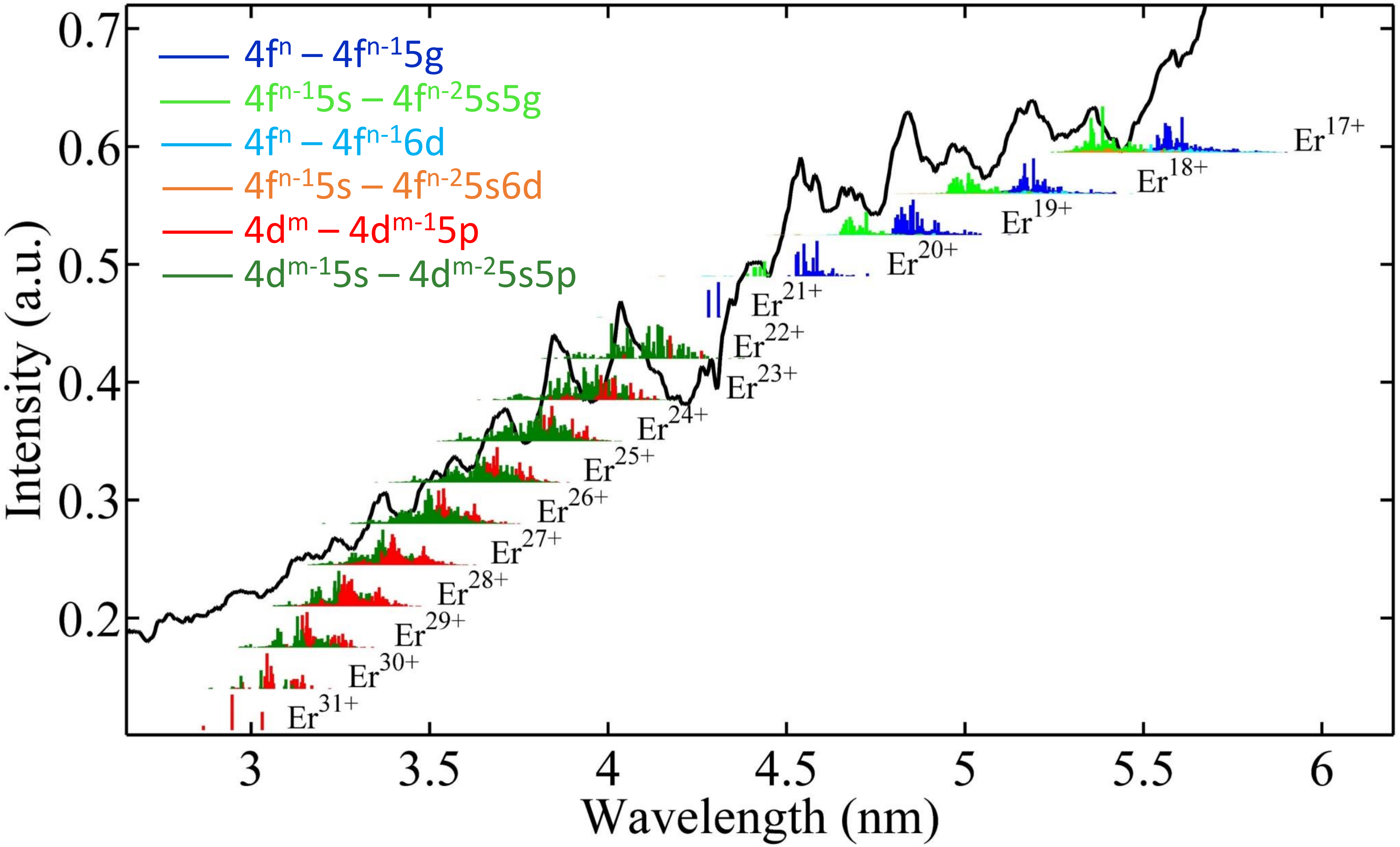
- Optimum scaling factors for Cowan code: 0.82, 0.92 and 0.96 → **Dramatic change over small ΔZ.**
- Scaling factor of 0.85 gives very accurate results for the heavier W^{38+} ion.

Tentative identifications

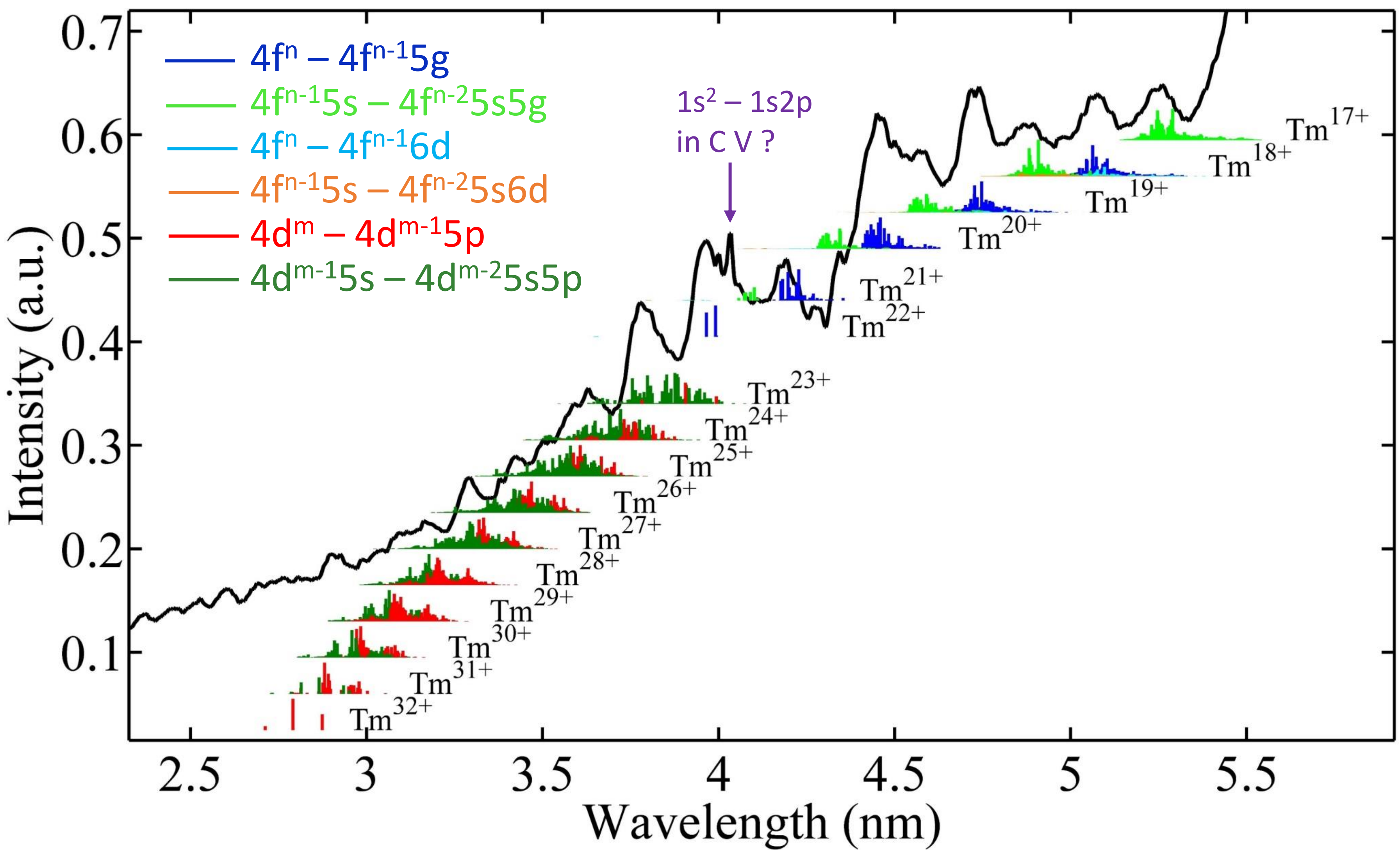
Moving to shorter λ….. Dy



Δn = 1 transitions: Er



Δn = 1 transitions: Tm



Conclusions

- Identified structure in the $\Delta n = 0$ arrays: Ag -, Pd - and Rh - like emission.

- Tentative identifications: Sr – like transitions $4d^2\ {}^3F_2 - 4d4f\ {}^3G_3$.

- Identified wavelength bands arising from 4f – 5g, 4d – 5p transitions and their corresponding 5s satellite arrays.

Publication

IOP Publishing — Journal of Physics B: Atomic, Molecular and Optical Physics

J. Phys. B: At. Mol. Opt. Phys. **50** (2017) 065006 (8pp) https://doi.org/10.1088/1361-6455/aa5e62

Analysis of soft x-ray emission spectra of laser-produced dysprosium, erbium and thulium plasmas

John Sheil[1,5], Padraig Dunne[1], Takeshi Higashiguchi[2], Domagoj Kos[1,3], Elaine Long[1], Takanori Miyazaki[1,2], Fergal O'Reilly[1], Gerard O'Sullivan[1], Paul Sheridan[1], Chihiro Suzuki[4], Emma Sokell[1], Elgiva White[1] and Deirdre Kilbane[1]

[1] School of Physics, University College Dublin, Belfield, Dublin 4, Ireland
[2] Department of Electrical and Electronic Engineering, Faculty of Engineering and Center for Optical Research & Education (CORE), Utsunomiya University, Yoto 7-1-2, Utsunomiya, Tochigi 321-8585, Japan
[3] Czech Technical University in Prague, Faculty of Nuclear Sciences and Physical Engineering, Brehova 7, 11519 Praha 1, Czech Republic
[4] National Institute for Fusion Science, 322-6 Oroshi-cho, Toki 509-5292, Japan

?

Multiconfiguration approach

Atomic State Function

Mixing coefficients

Configuration State Function

$$\Psi = \sum_i c_i \Phi_i$$

In the LS scheme, matrix elements written

$$H_{bb'}^{c_1,c_2} = \delta_{bb'} E_{av} + s\left\{\sum_k f_k F^k + \sum_{k'} g_{k'} G^{k'} + \sum_j d_j \zeta_j\right\}$$

$$H_{bb'}^{c_1-c_2,c_2-c_1} = s\left\{\sum_k r_d^k R_d^k + \sum_{k'} r_e^{k'} R_e^{k'}\right\}$$

where s is the *en-bloc* scale factor (*Ab initio* Hartree Fock: $s = 1$)

<ins>Highly mixed level</ins>

Composition very sensitive to choice of s. Scale factors used in Kato, *et al* → contribution of 4d4f $^3G_3$ CSF < another CSF – **change in level designation?**

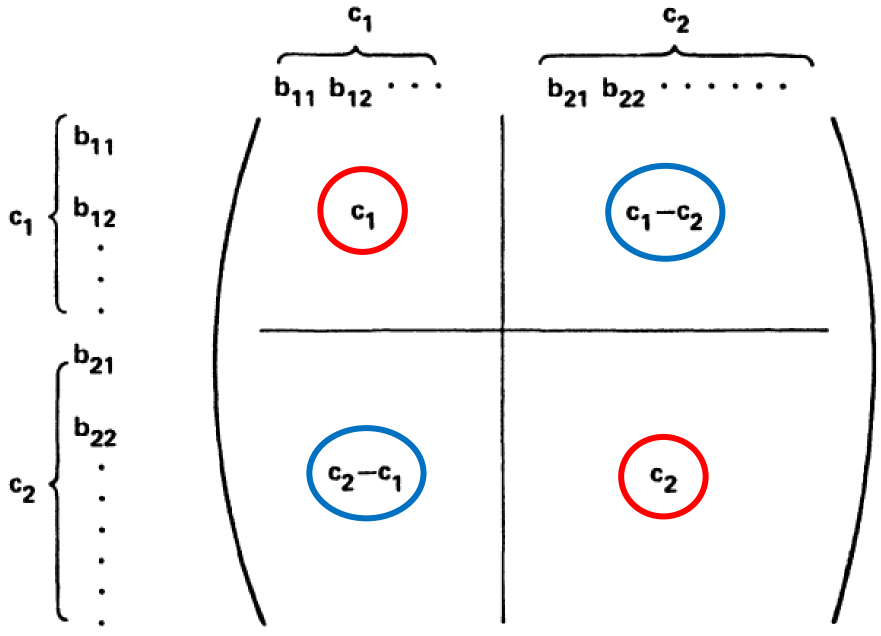


Fig. 13-1. General form of the Hamiltonian matrix (for any given value of J) in the two-configuration approximation. The basis states have been denoted by the labels b_{ij}, where i is the configuration number and j represents the serial number of the basis state within a configuration.

TASS, R.D.Cowan 1980

What has been studied?

Mainly **x-ray** spectra

Analysis of the x-ray spectrum emitted by laser-produced plasma of dysprosium

Gilad Marcus, Einat Louzon, Zohar Henis, and Shlomo Maman

Soreq Research Center, 81800 Yavne, Israel

Pinchas Mandelbaum

Jerusalem College of Engineering, Ramat Beth Hakerem, 91035 Jerusalem, Israel

2007

Classification of X-Ray Spectra from Laser Produced Plasmas of Atoms from Tm to Pt in the Range 6–9Å

P. Mandelbaum, M. Klapisch, A. Bar-Shalom and J. L. Schwob

Racah Institute of Physics, Hebrew University, 91904 Jerusalem, Israel

and

A. Zigler

Soreq Nuclear Research Center, 76400, Yavne, Israel

1983

The spectrum of highly ionized praseodymium and dysprosium from the Texas tokamak plasma in the 50–250-Å range

M. Finkenthal,[a] A. S. Lippmann, L. K. Huang, T. L. Yu, B. C. Stratton,[b] and H. W. Moos

Department of Physics and Astronomy, Johns Hopkins University, Baltimore, Maryland 21218

M. Klapisch, P. Mandelbaum, and A. Bar Shalom

Racah Institute of Physics, The Hebrew University, Jerusalem, Israel

W. L. Hodge, P. E. Phillips, T. R. Price, J. C. Porter, B. Richards, and W. L. Rowan

Fusion Research Center, The University of Texas, Austin, Texas 78712

1986

Recently…

EUV spectra of Rb-like to Ni-like dysprosium ions in an electron beam ion trap

Deirdre Kilbane[1], Gerald O'Sullivan[1], Yuri A. Podpaly[2], John D. Gillaspy[2], Joseph Reader[2], and Yuri Ralchenko[2,a]

[1] School of Physics, University College Dublin, Belfield, Dublin 4, Ireland

[2] National Institute of Standards and Technology, Gaithersburg, Maryland 20899, USA

2014

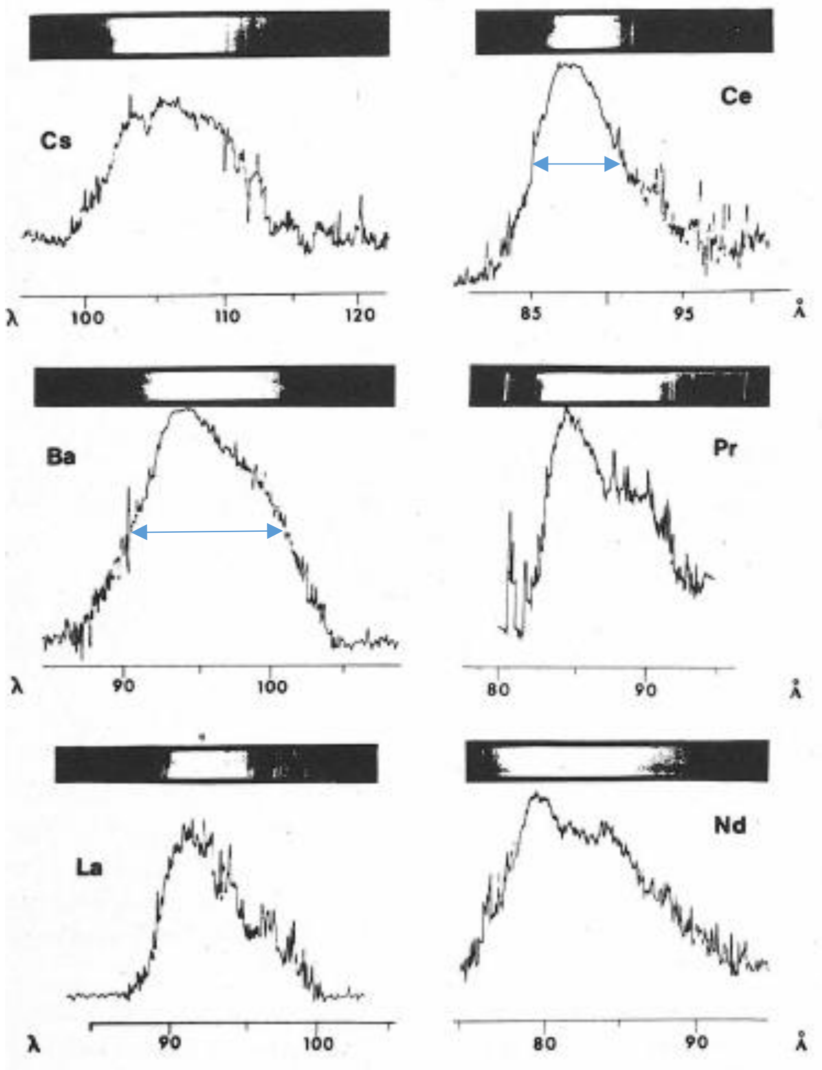
Measurements and identifications of extreme ultraviolet spectra of highly-charged Sm and Er

Y A Podpaly, J D Gillaspy, J Reader and Yu Ralchenko

National Institute of Standards and Technology, Gaithersburg, MD 20899, USA

2014

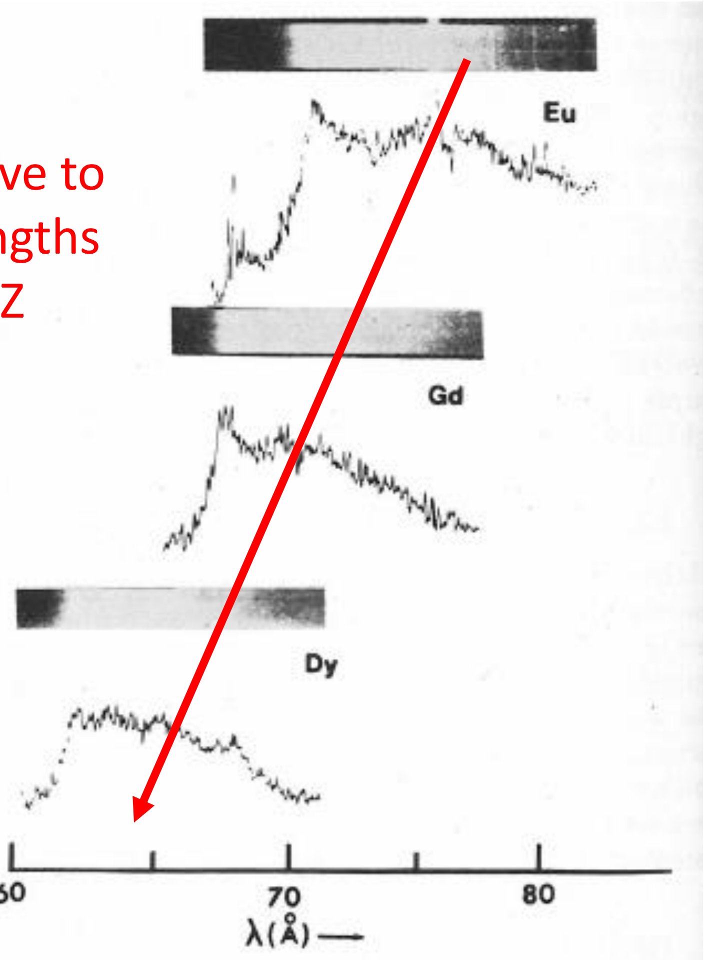
Early studies



O'Sullivan G and Carroll P K, J. Opt. Soc. Am. **71**, 227 (1981)

Narrow (9 – 18 eV) resonancelike emission attributed to 4d – 4f transitions

Resonances move to shorter wavelengths with increasing Z



CI between the upper $4p^5 4d^{n+1}$ and $4d^{n-1}4f$ configurations redistributes oscillator strength to the high energy end of the array → overlapping arrays in adjacent ion stages.